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Rapid Removal of Atmospheric CO_2 by Urban Soils

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Supporting Information

ABSTRACT: The measured calcium carbonate content of soils to a depth of 100 mm at a large urban development site has increased over 18 months at a rate that corresponds to the sequestration of 85 t of CO_2/ha (8.5 kg of CO_2 m^{-2}) annually. This is a consequence of rapid weathering of calcium silicate and hydroxide minerals derived from the demolition of concrete structures, which releases Ca that combines with CO_2 ultimately derived from the atmosphere, precipitating as calcite. Stable isotope data confirm an atmospheric origin for carbonate carbon, and ^{14}C dating indicates the predominance of modern carbon in the pedogenic calcite. Trial pits show that carbonation extends to depths of ≥1 m. Work at other sites shows that the occurrence of pedogenic carbonates is widespread in artificially created urban soils containing Ca and Mg silicate minerals. Appropriate management of fewer than 12000 ha of urban land to maximize calcite precipitation has the potential to remove 1 million t of CO_2 from the atmosphere annually. The maximal global potential is estimated to be approximately 700−1200 Mt of CO_2 per year (representing 2.0−3.7% of total emissions from fossil fuel combustion) based on current rates of production of industry-derived Ca- and Mg-bearing materials.

INTRODUCTION

There is a global commitment to reducing greenhouse gas emissions; the U.K. Government is currently committed to an 80% reduction by 2050 (against a 1990 baseline). A number of approaches envisage ways in which climate mitigation could be achieved practically. Pacala and Socolow estimated that 26 Gt of CO_2 $year^{-1}$ by 2050 would need to be removed from the atmosphere to compensate wholly for anthropogenic emissions. They propose that a number of individual mitigation approaches may potentially be used in unison to remove equally sized "stabilization wedges" of 4 Gt of CO_2 $year^{-1}$ each by 2050. This view was revised by Davis et al., who recognize that continued growth in annual emissions since 2003 required more wedges than previously considered and stress the importance of supporting energy technology innovation to achieve the required targets.[1,2]

Soils emit almost 275 Gt of CO_2 $year^{-1}$ as a consequence of plant root respiration and the mineralization of organic carbon, so they have an important influence on climate. The inorganic carbon content in soils is dominated by calcium and magnesium carbonate minerals.[3] During weathering, Ca and Mg silicate and carbonate minerals naturally react with dissolved carbon dioxide (CO_2) to increase local alkalinity. Given the right conditions, some of this dissolved CO_2 precipitates as secondary carbonate minerals in the soil. If the Ca or Mg in the carbonate is derived from silicate weathering, and weathering products, this process provides a means of effectively capturing and fixing atmospheric CO_2.[4−6] Examples of carbonation reactions are given below, using for illustrative purposes wollastonite, a natural calcium silicate (reaction 1), jennite, a hydrated cement mineral (reaction 2), and portlandite, a component of Portland cement (reaction 3):

$$CaSiO_3 + CO_2 + 2H_2O \rightarrow CaCO_3 + H_4SiO_4 \quad (1)$$

$$Ca_9Si_6O_{18}(OH)_6\cdot 8H_2O + 9H_2CO_3 \rightarrow 9CaCO_3 + 6H_4SiO_4 + 8H_2O \quad (2)$$

$$Ca(OH_2) + H_2CO_3 \rightarrow CaCO_3 + 2H_2O \quad (3)$$

The precipitation of carbonate minerals as a consequence of silicate weathering contributes to the stabilization of atmospheric CO_2 over geological time periods[4,7] and also contributes to the formation of pedogenic carbonates in both natural[8] and artificial soils[9] over much shorter time scales. Limited information is available for the formation of pedogenic carbonates in U.K. soils, with Rawlins[10] estimating the total inorganic carbon stock of soil (0−30 cm depth) to be 186 Mt

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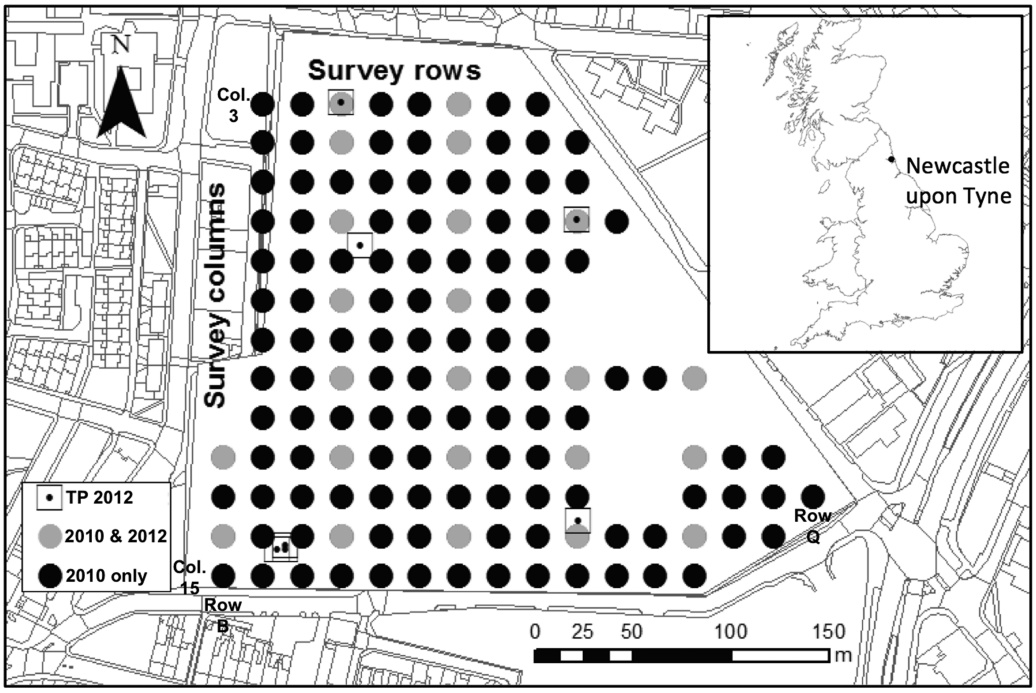

Figure 1. Location of study site, 2010[17] and 2012 sample points, and 2012 trial pit (TP) locations. Redrawn and updated version of figure taken from ref 17. Copyright 2012 Elsevier.

of C, around 5.5% of the estimated total soil carbon stock (organic and inorganic) across the U.K.

Work on the in situ weathering of natural and artificial silicates[9,11−20] has shown that artificial silicates and mineral wastes (such as construction and demolition waste, iron and steel slag, and mine tailings) in soil settings rapidly weather with the associated formation of carbonate minerals. This process is influenced by a number of physical and environmental factors, including small particle size and large surface area, poor crystallinity, and degree of exposure through proximity to the ground surface or position relative to the water table. In this way, artificial silicates have the global potential to capture 700−1200 Mt of CO_2 $year^{-1}$, representing 2.0−3.7% of contemporary global CO_2 emissions (currently global annual CO_2 emissions account for approximately 33 Gt of CO_2 $year^{-1}$), or one-sixth to one-third of a stabilization wedge.[1,21] Natural silicates present a carbon capture potential that is greater by orders of magnitude.[20]

Studies in urban soils[9,16] demonstrate a carbon capture potential of 12.5 kg of CO_2 (t of soil)$^{-1}$ $year^{-1}$ by this process, showing that formation of these pedogenic carbonate minerals in urban soils may be a significant and exploitable storage route for soil carbon. An increasing number of studies[9,12,14−19] have focused on the carbonation of artificial and waste minerals in this context, because of their abundance and ability to provide a readily available and accessible analogue for the intentional carbonation of natural minerals.

In the study presented here, we measured soil carbonate formation over 18 months, with accurate resampling of a subset of the original sample points achieved using high-resolution GPS. This is the first time that a study of this kind has directly measured changes in soil inorganic carbon content in urban soils over time. We quantifiably show that sequestration of CO_2 from the atmosphere can occur rapidly over months to years, even where sites are not intentionally managed for this purpose. Using stable and radiogenic isotope analysis, we unambiguously demonstrate the sequestration of modern atmospheric carbon dioxide. The results of this study suggest that the management of urban soils constitutes a promising and relatively inexpensive process for removing CO_2 from the atmosphere.

MATERIALS AND METHODS

Newcastle Science Central [World Geodetic System coordinates UK; 54.970406−1.626477 (WGS84)] is a 10 ha site that is currently being developed by Newcastle University and Newcastle City Council into a multiuse "science, business, living and leisure" complex.[22] The preexisting complex of brewery buildings was demolished in December 2007, at which point the site was completed with a surface layer (0.2−6 m thick) of "made ground" composed of material derived from demolition, including fines from the on-site production of crushed concrete secondary aggregates. This is typical of modern development practice, which frequently involves on-site crushing to recover a secondary aggregate product while retaining crusher "fines" on site as part of the soil. After demolition, the site remained undisturbed, allowing sparse growth of primary colonizing plants (e.g., *Buddleia* sp.), until early 2013 when ground stabilization work was performed to remove, replace, and compact all material to a depth of 20 m.

Soil samples were collected at 72 points on September 16, 2010,[17] and again at 23 of these points on March 13, 2012, located to 25 mm accuracy using Leica GPS equipment [Leica GS15 with a CS16 controller with a Smartnet RTK correction (Figure 1)]. At each location, the soil consisted of demolition rubble containing fragments of building materials grading into fines resulting from processing during demolition; 0.5−1.0 kg of this material was collected between 0 and 100 mm depth using hand tools and sieved to <2 mm on-site (eliminating large fragments of bricks and concrete, and representing a subsample of 30% of the total soil mass).[17] In parallel, seven trial pits, to investigate carbonate formation with depth, were excavated to a depth (where possible) of up to 3 m, on March 13, 2012, taking

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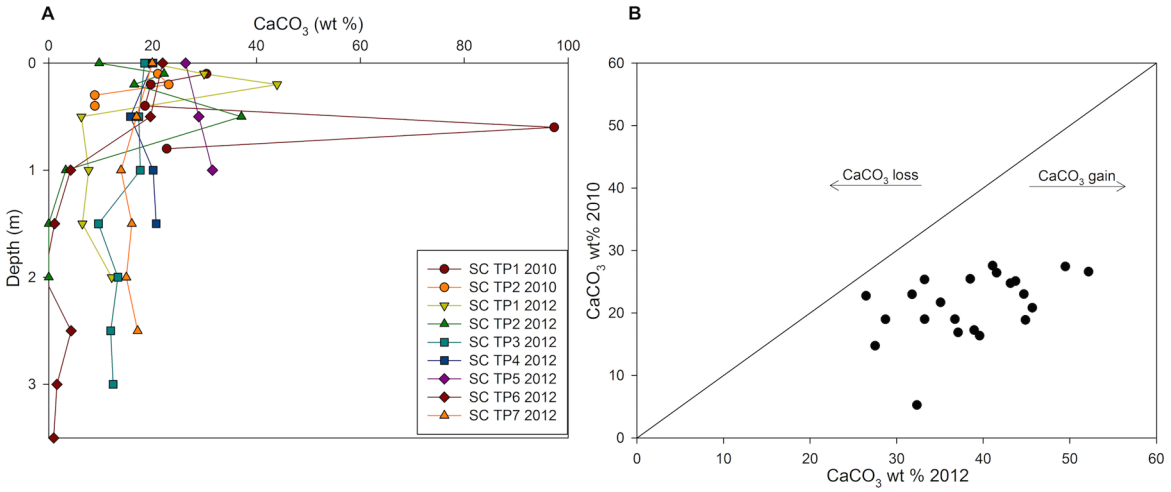

Figure 2. Calcium carbonate contents of urban soils determined by calcimeter analysis. (A) Variation in calcium carbonate content with depth from trial pit samples (N = 51). (B) Increase in $CaCO_3$ concentration from 2010 to 2012 from surface samples. The analytical error is within size of the symbol in both plots (comparative data 2010 vs 2012; N = 23).

samples of 0.5−1.0 kg at approximate intervals of 0.5 m. All samples were air-dried, crushed, and sieved to <63 μm prior to analysis.

Methods used are described fully in the Supporting Information, with full details of analytical accuracy and precision. Bulk carbonate was determined for 95 samples (72 for 2010, 23 for 2012) using an Eijkelkamp calcimeter (BS 7755-3.10:1995),[23] and organic (not reported here) and inorganic carbon contents were determined using a Leco CS-244 Carbon/Sulfur Determinator. Stable isotope ratios of C and O were determined for 18 (9 for 2010, 9 for 2012) samples by Iso-Analytical (Cheshire, U.K.) using a Europa Scientific 20-20 continuous-flow isotope ratio mass spectrometer (IRMS). Radiocarbon (^{14}C) measurement was performed on two samples (2010 sampling round) at the Scottish Universities Environmental Research Centre (SUERC) using an NEC Single Stage Accelerator Mass Spectrometer.[24,25] X-ray fluorescence (XRF) analysis was conducted on 14 samples (7 for 2010, 7 for 2012) at the University of Leicester Department of Geology using a PANalytical Axios Advanced XRF spectrometer. X-ray diffraction (XRD) analysis was performed on four samples (2 for 2010, 2 for 2012) at Newcastle University School of Chemical Engineering and Advanced Materials using a PANalytical X'Pert Pro Multipurpose Diffractometer fitted with an X'Celerator detector and a secondary monochromator. Thermogravimetry-differential scanning calorimetry coupled with quadrupole mass spectrometry (TG-DSC-QMS) was conducted for 6 samples (3 for 2010, 3 for 2012) using a Netzsch Jupiter STA449C TG-DSC system connected to a Netzsch Aeolos 403C QMS instrument.

RESULTS AND DISCUSSION

Surface soils (0−100 mm) were sampled systematically on two occasions 18 months apart, with very precise location (within 25 mm). All samples were analyzed immediately following collection (Table S1 of the Supporting Information). Where analyses were conducted on smaller subsets of surface samples, these were selected primarily to provide spatially even, distributed coverage across the study site.

The soil samples are similar in color and particle size distribution to those reported previously.[17] Soil pH was consistently high (>9.5) with values up to 11.7.[17]

Two methods of determination, calcimeter and Leco analysis (total carbon−organic carbon), give closely corresponding soil $CaCO_3$ contents. Calcimeter analysis reported bulk equivalent $CaCO_3$ contents of 39.4 ± 8.1 wt % (range of 26.5−61.4 wt %; analytical precision [standard error of the mean (SEM)] of 2010 samples, ±0.73 wt %; SEM of 2012 samples, ±0.21 wt %). Leco analysis reported an average inorganic carbon content of 4.1 ± 1.2 wt % (range, 3.0−7.1 wt %; SEM, ±0.48 wt %), corresponding to a $CaCO_3$ concentration of 38.9 ± 15.6 wt % (range, 25.2−59.1 wt %; equivalent analytical precision, ±4.0 wt %).

Figure 2A illustrates data from the depth to which trial pit samples could be taken during the sampling program; however, the total depth of material greatly exceeds this at some points across the site.[17] Trial pit samples to approximately 1 m depth have $CaCO_3$ contents varying from 0.0 to 97.3 wt %, with an average value of 19.1 wt % excluding a single outlier (97.3 wt % $CaCO_3$), which likely corresponds to inclusion of limestone aggregate (Figure 2A; variability with depth is due to changes in ground conditions). The average carbonate content for trial pits from approximately 0−3 m depth, calculated from Leco total inorganic carbon data, is 15.9 ± 9.7 wt % $CaCO_3$. $CaCO_3$ content does not vary with depth in a consistent manner across the site; nevertheless, the trial pit data generally show little variation with depth to ≥1.5 m. A number of the trial pits have anomalously high concentrations of $CaCO_3$ close to the surface (<1.0 m), and two exhibit a decline in concentration deeper than 1.0 m. These observations could indicate a carbonation front moving to greater depth with time. The observed variability can be attributed to heterogeneity in material composition and properties and to site-specific properties such as drainage patterns.

Thermogravimetry-differential scanning calorimetry coupled with quadrupole mass spectrometry (TG-DSC-QMS) and X-ray diffraction (XRD) analysis (Figures S1 and S2 of the Supporting Information) confirm the presence of calcite as the dominant calcium carbonate mineral and demonstrate the presence of portlandite [$Ca(OH)_2$].

Between sampling dates, the surface soil samples show a consistent and statistically significant increase in $CaCO_3$ content (Figure 2B). A Shapiro−Wilk test was conducted on the data, which found a non-normal distribution for 2010 and a

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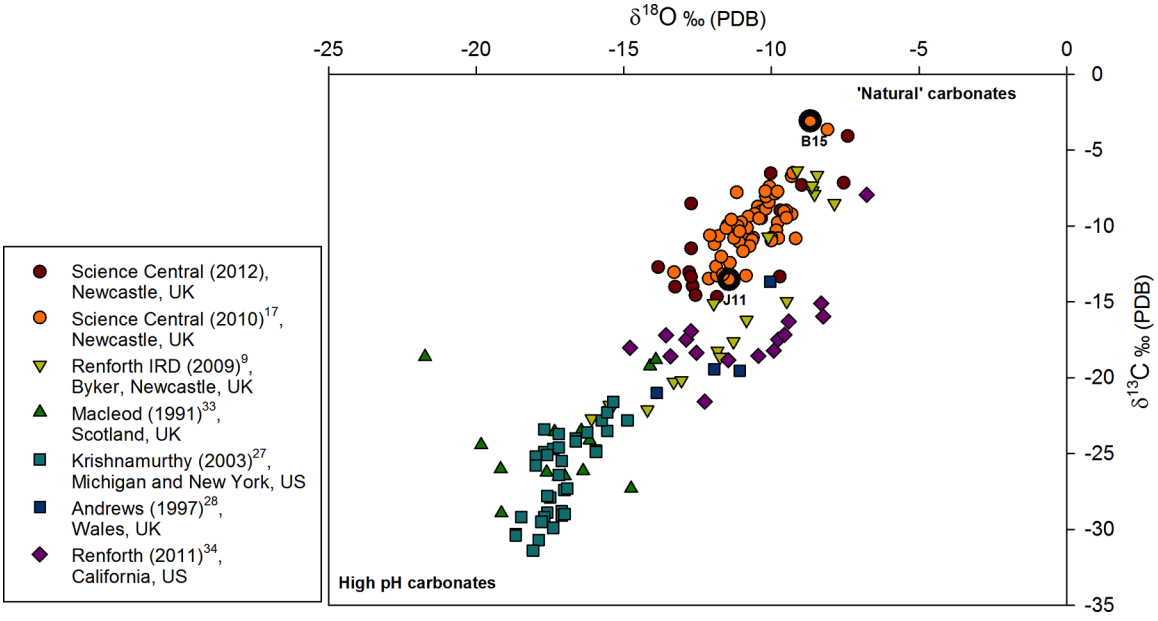

Figure 3. Stable isotopic data of samples from Science Central: $\delta^{18}O$ ‰ vs $\delta^{13}C$ ‰ (V-PDB) [repeatability error bars are within the point, in comparison with IRMS values from other sites (UK: 8, 26, 28; US: 25, 29)] (N = 185). ^{14}C sample points B15 and J11 are indicated.

normal distribution for 2012. Given this discrepancy, a nonparametric test was run (Wilcoxon Signed Rank Test, which is used to assess whether the rank of population mean values differs between related samples) producing a significance value of 0.000, allowing the rejection of the null hypothesis (at 95% confidence) that the difference in the samples was due to chance. The starting composition of the material with respect to $CaCO_3$ is not known but can be assumed to be small (but non-zero) because of the possible inclusion of limestone aggregate or concrete carbonation occurring over the life of the now demolished structures. Overall, the $CaCO_3$ content to 100 mm depth over 18 months (2010−2012) increases from 22.0 to 39.0 wt %. The range of the entire sample data is 5.3−43.0 wt % $CaCO_3$ in 2010 and for the repeat samples 26.5−61.4 wt % $CaCO_3$ in 2012. Standard deviations are higher (8.3 wt %) in 2012 (6.9 wt % in 2010), which may reflect variable progress of the carbonation reaction occurring in a geochemically heterogeneous environment.

A number of studies[26−28] have illustrated that the stable isotope signatures of carbon and oxygen can be used to assess sequestration of CO_2 in recently formed pedogenic inorganic carbonates. Nonequilibrium stable isotope signatures are common in carbonate minerals formed from high-pH brines. Thus, nonequilibrium isotope fractionation must be considered in studies of weathering and carbonation of portlandite, $Ca(OH)_2$, which releases hydroxyl ions into solution (hydroxylation), thereby increasing the pH. Hydroxylation occurs at high pH (>11.5),[26] in environments corresponding to those dominated by the weathering of artificial minerals in construction materials, as at Science Central. The C isotope signature is governed by diffusion of CO_2 into an aqueous solution and subsequent precipitation of $CaCO_3$. The rate of hydroxylation exerts the greatest influence on the isotope signature of high-pH solutions and can be assigned a large role in determining the isotopic composition of carbonates formed from the weathering of artificial minerals in soil settings. Stable isotopic analysis of secondary carbonate minerals forming under ambient conditions that are relevant to soils confirms that a significant proportion of the carbonate carbon has been sequestered from the atmosphere in this way.[29]

C and O isotope data for Science Central have $\delta^{18}O$ values between −10.0 and −13.8‰ and $\delta^{13}C$ values between −6.5 and −14.6‰ (all V-PDB; 2010 samples, SEM of ±0.03‰ for $\delta^{13}C$ and ±0.06‰ for $\delta^{18}O$; 2012 samples, SEM of ±0.04‰ for $\delta^{13}C$ and ±0.03‰ for $\delta^{18}O$). Between the two sampling dates, the mean $\delta^{13}C$ values change from −9.9 to −12.6‰ and $\delta^{18}O$ from −10.7 to −12.6‰. The shift toward more negative values is expected if the carbonate present includes a higher proportion produced following hydroxylation. The data shown in Figure 3 represent mixtures of $CaCO_3$ derived from geological (lithogenic) sources (such as limestone aggregate in construction materials) and carbonate that originates from hydroxylation of CO_2 at high pH,[9] forming a linear plot representing mixing between lithogenic and pedogenic carbonates.[9] Similar arrays of data are reported for mortar[27] and carbonate precipitates from the underside of concrete structures.[33]

A mixing line is superimposed on the data,[17] which assumes a lithogenic end member value for $\delta^{18}O$ of −0.0‰, a $\delta^{13}C$ of −0.0‰, and a hydroxylation (high-pH) end member $\delta^{18}O$ of −20.5‰ (assuming a meteoric water $\delta^{18}O$ of −7.8‰ vs SMOW) and a $\delta^{13}C$ of −25.3‰[9,17,26,28,30,31] (Figure S3 of the Supporting Information). Other studies consider mixing between lithogenic carbonate and a "biogenic" end member, with a $\delta^{18}O$ of −6.8‰ and a $\delta^{13}C$ of −8.0‰, attributed to photosynthesis.[9,32] As the soils investigated in this study have little plant cover, and the observed range of $\delta^{18}O$ and $\delta^{13}C$ extends to more negative values, it is assumed that photosynthetic inputs to newly formed soil carbonate are negligible. Thus, on the basis of mixing between lithogenic and pedogenic carbonate formed at high pH, proportions of each component vary from 42 to 80% lithogenic and from 58 to 20% hydroxylation (mean hydroxylation of 50%).

The data shown in Figure 3 represent mixtures of $CaCO_3$ derived from geological (lithogenic) sources and carbonate that is assumed to have originated more recently from the

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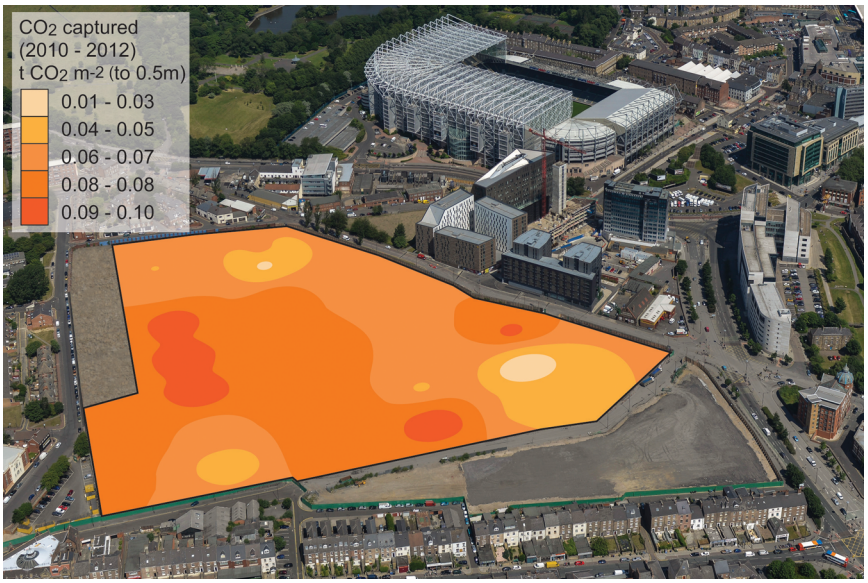

Figure 4. Observed carbon capture at Science Central, Newcastle (N = 23). Contour map plotted using ArcGIS 10. Overlay created in Adobe Photoshop and Illustrator.

hydroxylation of CO_2 at high pH.[9] ^{14}C measurements (two samples selected from the extremes of the observed range of the $\delta^{13}C$ data) suggest that a significant proportion of the C present in carbonates originates from the modern atmosphere (i.e., nonlithogenic sources). A value of 30.4 ± 0.1% Modern Carbon was found for a sample with a $\delta^{13}C$ of −3.13‰ (B15) and 80.7 ± 0.4% Modern Carbon for a sample with a $\delta^{13}C$ of −13.55‰ (J11; points are indicated in Figure 3). Full ^{14}C data are listed in Table S2 of the Supporting Information. This is consistent with an increased level of incorporation of carbon from the modern atmosphere in samples that show the most reaction.

Variation in $CaCO_3$ content with depth depends on ground conditions, as noted in previous sections. In general, homogeneously crushed demolition material was found to depths of at least 1 m, extending deeper than this in some cases. On the basis of the observed change between 2010 and 2012 in $CaCO_3$ content of surface samples from 370 to 660 t ha^{-1} (0.037−0.066 t m^{-2}), the rate of precipitation of calcium carbonate in the top 100 mm of soil is estimated to be 16 t ha^{-1} (0.0016 t m^{-2}) per month, or 192 t ha^{-1} (0.0192 t m^{-2}) annually. This corresponds to the removal of carbon from the atmosphere equivalent to 85 t of CO_2 ha^{-1} (0.0085 t of CO_2 m^{-2}) annually, or an annual increase in soil C content of 23 t of C ha^{-1} (0.0023 t of C m^{-2}).

The measured soil inorganic carbon stock to 100 mm depth increased from 44 to 79 t of C ha^{-1} (0.0044−0.0079 t of C m^{-2}) in 18 months, approaching values similar to those reported for organic carbon in arable land [84 t of C ha^{-1} (0.0085 t of C m^{-2})].[35] The rate of increase inferred from the observed change at Science Central exceeds estimates for soil carbon increases in rural land use [e.g., 3.63 t of C ha^{-1} $year^{-1}$ (0.363 kg of C m^{-2} $year^{-1}$) through establishment (for example) of new woodlands].[36] It is likely that the observed rate will change with time, but it is not possible to measure changes at the site since 2012 because of more recent construction activity.

Soil carbon stocks are conventionally reported to greater depths than 100 mm.[10] Figure 4 summarizes CO_2 removal, extrapolating to 0.5 m depth on the basis of the consistent $CaCO_3$ contents to that depth from trial pit samples (see Figure 2A) The extrapolation in Figure 4 is based upon trial pit data to 0.5 m and analytical data, assuming a contemporary mean $CaCO_3$ content of 20 ± 12.3 wt % (mean $CaCO_3$ content from trial pits to 0.5 m and surface samples of 19.5 wt %) and assuming that the proportion of carbonate newly formed (shortly prior to or since demolition) through hydroxylation is 50% overall.

The calcium required for the carbonation reaction derives from two sources. First, dissolution of the cement mineral portlandite [$Ca(OH)_2$] can provide readily available Ca. The portlandite content of the samples was not quantified but is typically around 15% in hydrated cement (∼5% in concrete).[37] Second, dissolution of artificial Ca-bearing silicates (amorphous or poorly crystalline phases; part of the cement clinker) occurs as cement weathers within the soil. An estimate of the rate of weathering of the silicate components of the soil has been made using the change in bulk composition determined by XRF (Table S3 of the Supporting Information), which shows a negative correlation between SiO_2 and CaO, consistent with the formation of calcite and proportionate reduction in the silicate mineral fraction. However, once normalized to TiO_2 (assuming Ti to be immobile during weathering),[38] the loss of SiO_2 exceeds that corresponding to dilution by newly formed carbonate minerals. The observed loss of 20% of the SiO_2 content is equivalent to a weathering rate of $10^{-12.7}$ mol of Si cm^{-2} s^{-1} (normalized to a nominal mineral surface area of 1 m^2 g^{-1}, conservatively estimated from particle size distribution[17] and assuming 1 Mt of material on-site[22]) or 16000 t of Si km^{-2} a^{-1} (when normalized to the site area). It is assumed that Si is removed from the site in drainage waters, although some may have been retained in the formation of clay minerals (identified by XRD analysis). This is several orders of magnitude slower than dissolution rates for hydrated cement minerals determined through laboratory experiments ($10^{-8.0}$ mol of Ca cm^{-2} s^{-1})[17] but is 2 orders of magnitude faster than the greatest weathering rates for natural catchments (e.g., a global average of 24 t km^{-2} $year^{-1}$,[33] or 100s t km^{-2} $year^{-1}$ for tropical volcanic arcs).[39,40] Enhanced SiO_2 removal is consistent with the high pH of this

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anthropogenic soil and may explain the elevated weathering rate measured from rivers draining urbanized catchments.[41]

The significance of the results reported here is the demonstration of the very rapid and substantial rate of removal of CO_2 from the atmosphere by carbonation reactions that take place within soils that contain reactive calcium minerals. Similar observations have been found at other sites, where the rate of CO_2 removal has been estimated on the basis of the assumption that calcium carbonate content in soil was zero at the time of demolition[9] or soil formation.[14] In urban soils, the dominant calcium silicate flux to soil is by incorporation of demolition materials. Carbonation of artificial cement replaces CO_2 lost during manufacture, which involves calcining limestone (globally 5% of anthropogenic emissions),[42] thus closing one loop of the carbon cycle but not compensating for CO_2 produced by combustion of fuel used in the calcining process. Science Central represents a single 10 ha site but is typical of modern development practice that involves on-site crushing of materials and the retention of crusher "fines" on-site as part of the soil. Urban land in the U.K. represents 7% (or 1.7 million ha) of the country's 25 million ha area.[43] If only 12000 ha (or 0.7%) of urban soils in the U.K. were managed in a way similar to that of Science Central, they could have the potential to remove 1 Mt of CO_2 annually over the period in which they are sequestering carbon dioxide, contributing to the requirement for the U.K. carbon account for 2050 to be at least 80% lower than the 1990 baseline.[44]

There are several considerations around the effective management of these sites to maximize carbon capture, including the rate and thickness at which materials are emplaced, how long the site remains undeveloped, and the manner in which it might be covered by buildings or by landscaped topsoil. Science Central is typical of redevelopment land, where demolition materials are emplaced to variable depth (1 m to tens of meters) on sites that may remain undeveloped for 5 or more years. Carbonation appears to decrease with depth, likely due to the isolation of materials from the atmosphere; therefore, management that increases atmospheric exposure (e.g., through appropriate planting that takes CO_2 to depth via plant root systems and associated metabolism) is likely to increase carbonation rates throughout the site.[14,45,46] There is evidence that this carbon capture process occurs at other sites where topsoil has been emplaced.[9] If the soil is covered by a building or impermeable pavements it will be isolated from the atmosphere, due to a "capping" effect. By analogy with natural systems, carbonate material formed while sites are undeveloped is likely to be stable and remain in the soils during and after redevelopment has taken place.[47]

Globally, appropriate management of land as part of the development or reconstruction of sites where concrete structures have been demolished or destroyed should be considered as a way of partly compensating for emissions associated with the loss of CO_2 from firing carbonate raw materials (around 50% of the total CO_2 emissions of a kiln).[48] However, the cement industry will have to rapidly decarbonize in the coming decades to meet emission reduction targets (including wide-scale deployment of carbon capture and storage of kiln flue gases). The postuse recarbonation of cement at the end of life of a structure, through soil processes, could result in a net negative carbon life cycle if combined with mitigation at the kiln. Approximately 3×10^9 t of cement is produced annually, emitting approximately 1.5×10^9 t of CO_2 from chemical decarbonation.[49] Given the average building lifespan is around 50 years, this material could become available by midcentury, representing a substantial carbon capture opportunity for the industry.

Although reported here for urban soils with artificial mineral inputs, calcite precipitation has also been observed in artificial soils prepared by blending natural calcium silicate rocks with compost.[14] In the long term, a proportion of the calcite may dissolve, contributing to bicarbonate in groundwater and surface water (fluxes similar to those from outcropping natural carbonate-cemented sandstones). The potential for carbon capture as calcite precipitated in soils with added calcium silicates has the potential to provide a passive carbon dioxide removal mechanism, analogous to the use of reed beds to clean contaminated surface waters.

■ ASSOCIATED CONTENT

Supporting Information

Additional methods, data, and figures. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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